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FORMATION AND CONTROL OF NO EMISSIONS FROM COAL-FIRED SPREADER-STOKER BOILERS

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16. ABSTRACT

The paper describes results of a study on the formation and control of nitrogen oxide (NO) in coal-fired spreader-stoker systems. (Stoker-coal-fired furnaces are significant in terms of coal consumption and environmental impact; however, they have received little research attention.) Three scales of experimental equipment were used to define the evolution and oxidation of fuel nitrogen in the fuel suspension phase, the conversion of fuel nitrogen during fixed-bed combustion, and the coupling between the two combustion phases. Results indicate that NO emissions from spreader-stoker coal-fired furnaces are the result of relatively high conversions of fuel nitrogen evolved from particles of < 0.1 in. diameter in the suspension phase and low conversion of fuel nitrogen during fixed-bed combustion. In the suspension phase, nitrogen is evolved at about the same rate as carbon is oxidized. Local oxygen availability is the primary control parameter for both combustion phases. Minimum overall fuel nitrogen conversions of 6 percent were achieved in the pilot scale facility by controlling the stoichiometry in both combustion zones.

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Nitrogen Oxide (NO)	Nitrogen	Stationary Sources	07B	
Coal	Carbon		21D	
Combustion			21B	
Boilers			13A	
Stokers				

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ABSTRACT

Stoker-coal-fired furnaces are significant in terms of coal consumption and enviromental impact; however, they have received little research attention. This paper describes the results of a study on the formation and control of nitric oxide in coal-fired spreader-stoker systems. Three scales of experimental equipment were used to define the evolution and oxidation of fuel nitrogen in the fuel suspension phase, the conversion of fuel nitrogen during fixed-bed combustion, and the coupling between the two combustion phases.

The results indicate that NO emissions from spreader-stoker coal-fired furnaces are the result of relatively high conversions of fuel nitrogen evolved from particles of less than 0.1 in. diameter in the suspension phase and low conversion of fuel nitrogen during fixed-bed combustion. In the suspension phase, nitrogen is evolved at approximately the same rate as carbon is oxidized. Local oxygen availability is the primary control parameter for both phases of the combustion. Minimum overall fuel nitrogen conversions of 6% were achieved in the pilot scale facility by controlling the stoichiometry in both combustion zones.

INTRODUCTION

In recent years considerable research has focused on pulverized coal burning; however, many current industrial coal units are stoker-fired, and these systems have not received much attention. Stoker-fired boilers are significant in terms of both coal consumption and contributions to national NO_x and SO_x emissions. Almost 20% of the coal consumed in the U.S. is burned in stoker systems. They are the single largest source of combustion-generated particulate emissions and a major source of NO_x and SO_x emissions.[1]

The spreader stoker is the most common large mechanical stoker. In this system a multiblade overthrow rotor projects large coal particles (80% > 8 mesh; max size = 1.25 in.) onto a moving or vibrating grate. The smaller particles burn in suspension while the heavier pieces fall to the grate for combustion in a thin (2-6 in.) bed. Coal heat release rates are approximately 750,000 Btu/hr/ft^2.

Recent pilot and field tests have partially demonstrated the potential of combustion modifications for NO_x control in stoker-fired boilers. Tests on a 25,000 lb/hr steam spreader stoker indicated that decreased overall excess air resulted in lower NO_x emissions.[2] Field results for large-scale industrial stokers have shown NO_x emissions to be insensitive to changes in overfire-air percentage and to increase with increasing load.[3] Laboratory experiments in a 180 mm pot furnace (batch operation) have suggested that the oxidation of volatile fuel nitrogen may be of major importance in stoker systems, particularly in the early portion of the burning.[4]

This paper summaries an EPA-funded program to study the formation and control of nitric oxide (NO) in coal-fired spreader stokers. In the experiments only NO was routinely measured since the other major species,

nitrogen dioxide (NO_2), was normally a small fraction of the total NO_x emission. The approach was primarily experimental; three types of laboratory furnaces were utilized to investigate the suspension burning, the bed combustion, and the overall process. Exhaust and in-flame species and temperature measurements were made with bituminous and lignite coals to define the influence of combustion conditions on NO emissions.

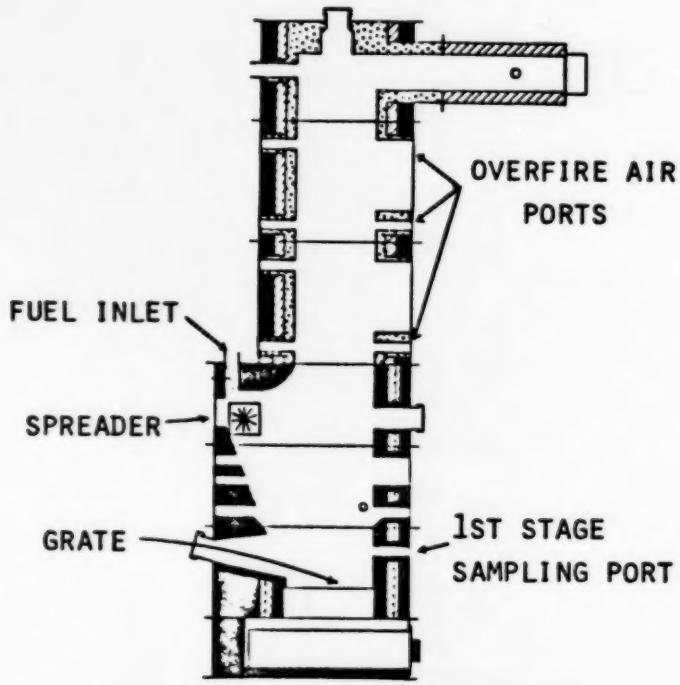
EXPERIMENTAL

Model Spreader-Stoker Furnace

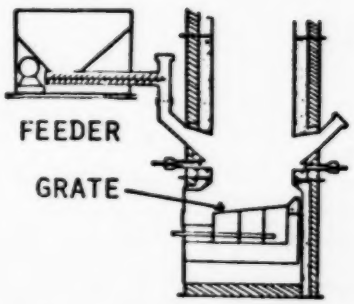
The primary experimental facility used in this work was the 750,000 Btu/hr model spreader-stoker furnace (MSF) shown in Figure 1. This eight-section, refractory wall unit had a 1 ft^2 dumping grate and the vertical dimensions were scaled approximately 1:1 with commercial practice. Coal was fed at 60 lb/hr onto a rotating, multivane spreader by a metering auger. The variable speed spreader, located 32 in. above the stainless- steel grate, distributed the coal uniformly across the bed. All sections contained multiple sampling ports, and overfire-air slots. All air streams were at ambient temperature.

Tower Furnace

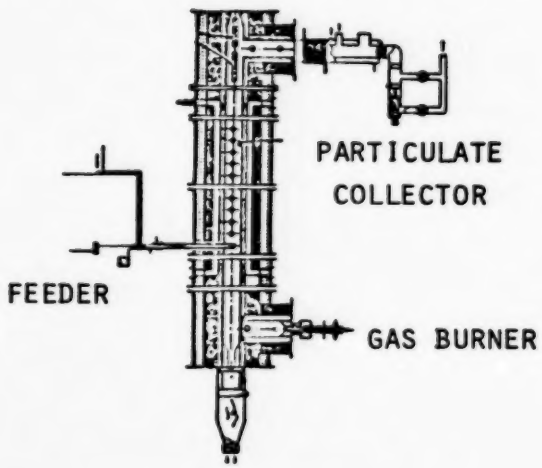
The 70,000 Btu/hr tower furnace (Figure 1B) was utilized to investigate suspension phase combustion. This refractory wall combustor[5] was 6 in. in inner diameter, 8.5 ft long, and contained provision for auxiliary heating (by firing natural gas in annular channels around the main chamber) to maintain constant wall temperature. Sized coal particles were fed into the furnace center through a stainless-steel water-cooled injector 32 in. above the bottom. A high-intensity gas burner, attached to the horizontal extension at the furnace bottom, produced hot combustion products which flowed vertically upward to simulate the combustion products leaving an actual stoker bed. Particles falling to the furnace bottom were collected in a water-quenched trap. Unburned particles reaching the flue were collected in a water-quenched, 200-mesh screen system.



A. MODEL STOKER FURNACE



C. FIXED-BED FURNACE



B. TOWER FURNACE

Figure 1. Experimental Furnace Facilities

Fixed-Bed Furnace

The 300,000 Btu/hr fixed-bed furnace (Figure 1C) was 11 in. by 15 in. inside, had a 0.75 ft^2 grate, and was used for detailed investigation of bed combustion.[6] The composite refractory walls contained multiple overfire-air slots, large observation windows, and in-flame probe ports. Coal was continuously metered onto the grate from above to simulate the bed-feeding process in a spreader stoker. Suspension phase burning of the fines was essentially eliminated by sizing the coal to 0.25 by 1 in. The ambient temperature combustion air was distributed between the three-section underfire-air plenum and two levels of overfire-air ports.

Analytical Measurements

Exhaust gas samples were withdrawn with a water-cooled stainless-steel probe and pumped to the continuous instrumentation which included a chemiluminescent NO analyzer, a paramagnetic O_2 analyzer, and an NDIR analyzer for CO and CO_2. In-flame HCN samples were withdrawn through a controlled-temperature (250°F) probe and analyzed with a HP 5840 gas chromatograph containing an NPD. NH_3 was sampled with an acid spray probe, collected in a series of bubblers, and analyzed with a gas sensing electrode.[7]

Fuels

Table 1 shows the ultimate and proximate analyses of each of the coals used in this study. The coals were sized for the tower furnace (0.0 - 0.063 in.) and the fixed- bed furnace (0.25 - 1.0 in.) as indicated. In the model spreader stoker the coals were burned on an as-received basis.

Table 1.

Fuel Analysis

PROXIMATE ANALYSIS	UTAH	NORTH DAKOTA LIGNITE	INDIANA	VIRGINIA
Moisture	6.20	29.90	2.96	4.86
Ash	9.20	6.76	14.15	13.48
Volatile Matter	37.62	29.74	33.45	27.28
Fixed Carbon	46.98	33.60	49.44	54.38
ULTIMATE ANALYSIS				
Moisture	6.20	29.90	2.96	4.86
Carbon	66.93	44.74	61.79	68.16
Hydrogen	4.65	2.96	4.16	4.26
Nitrogen	1.11	0.67	1.24	1.76
Sulfur	0.33	0.37	2.17	1.73
Oxygen	11.58	14.60	13.53	5.75
Ash	9.20	6.76	14.15	13.48
Fuel Constituent (%, daf)[1]				
Nitrogen	1.31	1.06	1.10	2.16
Sulfur	0.39	0.58	2.60	2.12
Particle Size (in.)	0.25 - 1	0.25 - 1	0.0 - 0.063	0.0 - 0.063
Calorific Value (Btu/lb) (as rec'd)	11,571	7,573	10,829	12,206
Calorific Value (wet/mmf)[2] (Btu/lb)	12,743	8,122	12,610	14,100
ASTM Rank[3]	HVC Bituminous	Lignite A	HVC Bituminous	HVA Bituminous

[1] daf: dry, ash free
[2] mmf: Mineral matter free
[3] HV: high volatile

SUSPENSION COMBUSTION

This section of the paper summarizes the tower furnace results which show the importance of suspension phase particle combustion. The objectives of these studies were to define those particle sizes which experienced significant reaction in the overthrow phase and to establish the fate of the nitrogen in those particles.

Combustion Characteristics

Figure 2 summarizes the influence of particle size, mean-free-stream oxygen, and thermal environment on the percent of inlet carbon oxidized in the suspension phase for Indiana coal. Particle size was the most important parameter studied. At standard commerical operating conditions (6% O_2) more than 90% of the particles of less than 0.063 in. diameter were entrained in the upward flow and more than 60% of this feed burned. However, less than 20% of the particles with diameters between 0.063 and 0.111 in. burned and nearly all particles of greater than 0.11 in. diameter fell to the furnace bottom essentially unreacted. Increasing the mean-free-stream-oxygen concentration (the average between the furnace top and bottom) increased CO_2 production due to both increased entrainment in the upward gas flow (resulting from higher vertical velocities) and increased oxygen partial pressure. Decreasing the reaction temperature by approximately 200°F (via decreased auxiliary firing) decreased the suspension burning by about 15%.

Fate of Coal Nitrogen

Figure 3 shows the fate of the Indiana coal nitrogen as a function of the mean oxygen concentration for the 1900°F, d < 0.063 in. case. Nitrogen

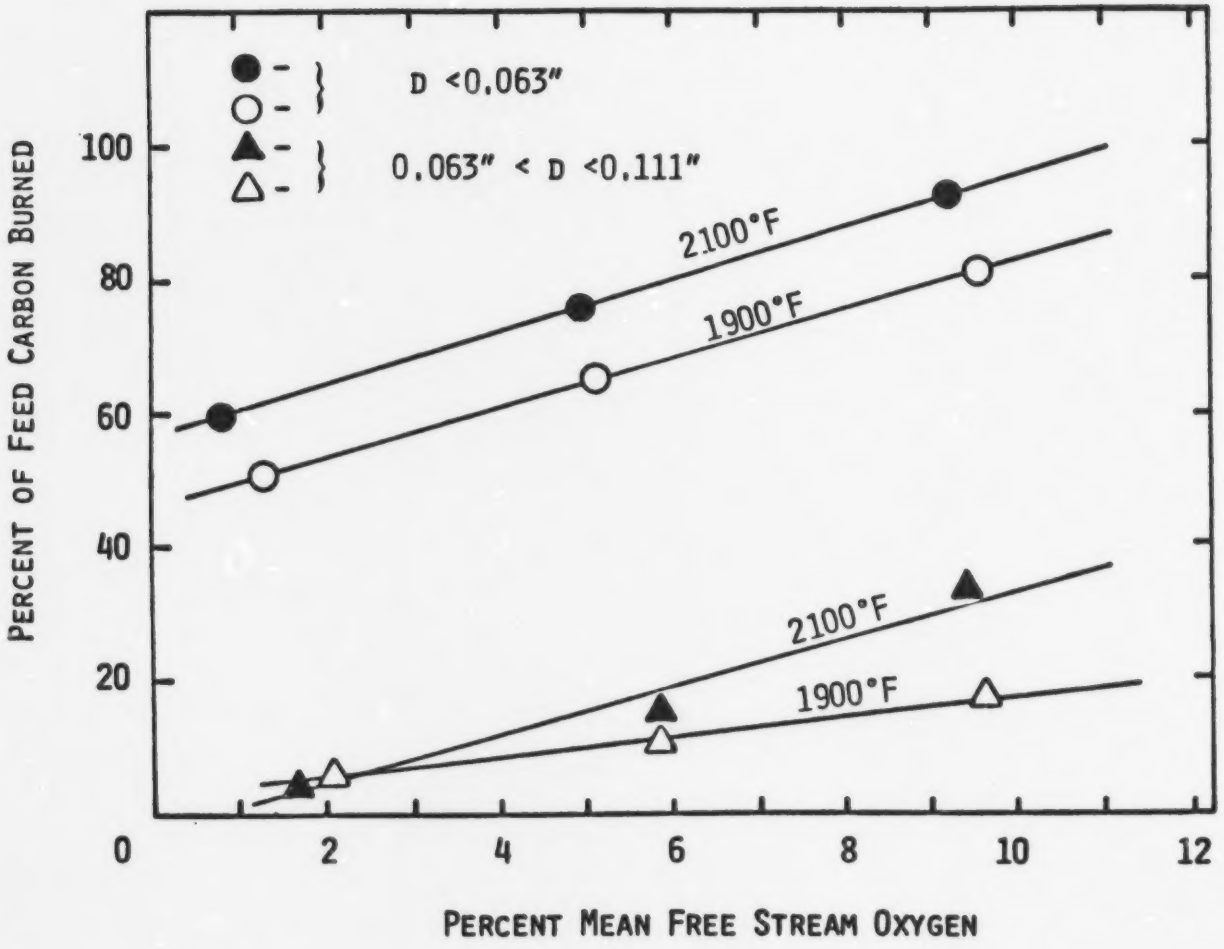


Figure 2. Suspension Combustion Results (Indiana Coal, Tower Furnace)

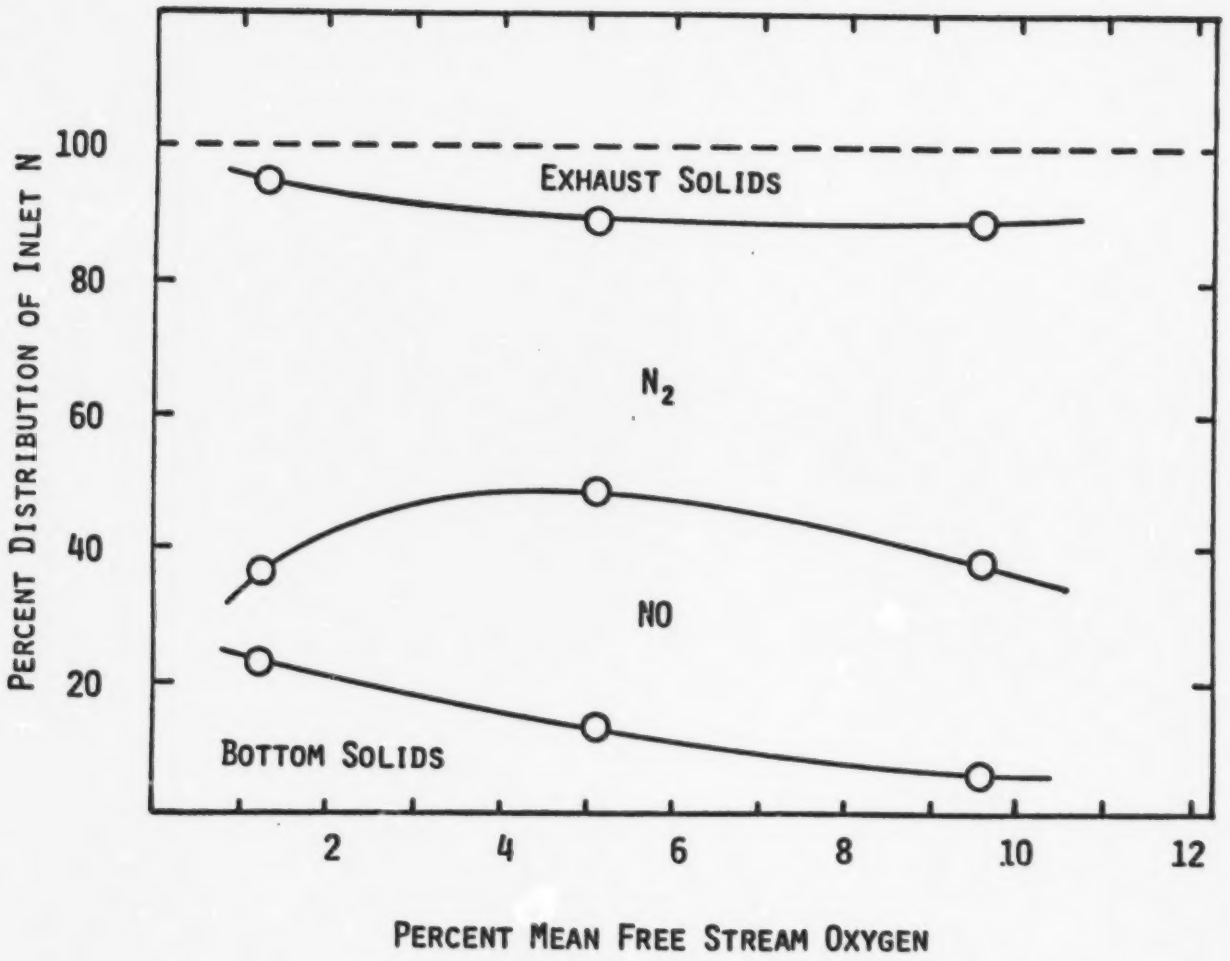


Figure 3. Fate of Inlet Nitrogen (Indiana Coal, 1900°F, d <0.063 in., Tower Furnace)

retention in the exhaust and bottom solids was determined from the composition and mass of the total solids collected at each location. NO formation was determined by measuring the NO concentrations with and without the coal feed. Increasing the mean oxygen decreased the fraction of nitrogen reaching the bed and slightly increased that carried into the exhaust; both effects are primarily due to increased particle entrainment. NO formation decreased significantly at low mean-free-stream-oxygen concentrations as expected from pulverized coal data.[8]

Figure 4 shows the fate of the nitrogen remaining in the solid phase relative to the solid phase carbon in the char (normalized with respect to the nitrogen-to-carbon ratio in the unburned coal). Particles which fell to the bottom had burned only slightly (the hydrogen content of the solid was still significant) and exhibited some nitrogen enrichment. Thus, the evolution of nitrogen slightly lagged the evolution and oxidation of the early hydrocarbon volatiles, and this is in agreement with inert pyrolysis studies.[9, 10] However, the N/C ratio in the exhaust solids was less than that in the raw coal, demonstrating that the final nitrogen compounds reacted preferentially to the most refractory char species. Since the exhaust solids exhibited behavior opposite to that of the bottom solids, and since their relative masses were approximately equal for the 0.063 in. particles, the data shown in Figure 4 indicate that the overall nitrogen evolution rate was near that of carbon. The exhaust data also showed that decreasing particle size and increasing bulk gas temperature both increased fuel nitrogen evolution.

Figure 5 summarizes the effects of the combustion parameters on the % of the evolved fuel nitrogen which was ultimately converted to NO. Conversions were relatively high (ca. 40%) and essentially independent of the

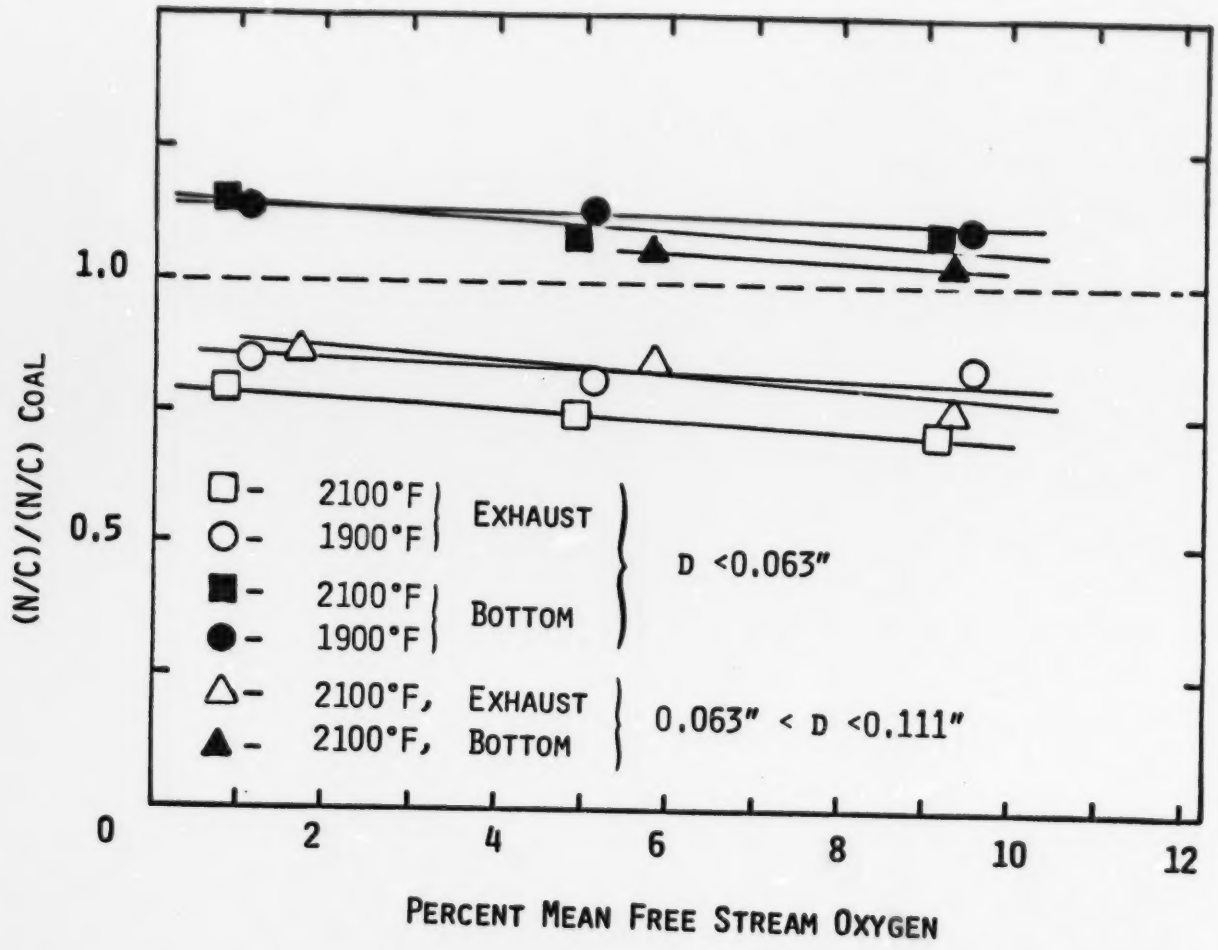


Figure 4. Distribution of Solid Nitrogen (Indiana Coal, Tower Furnace)

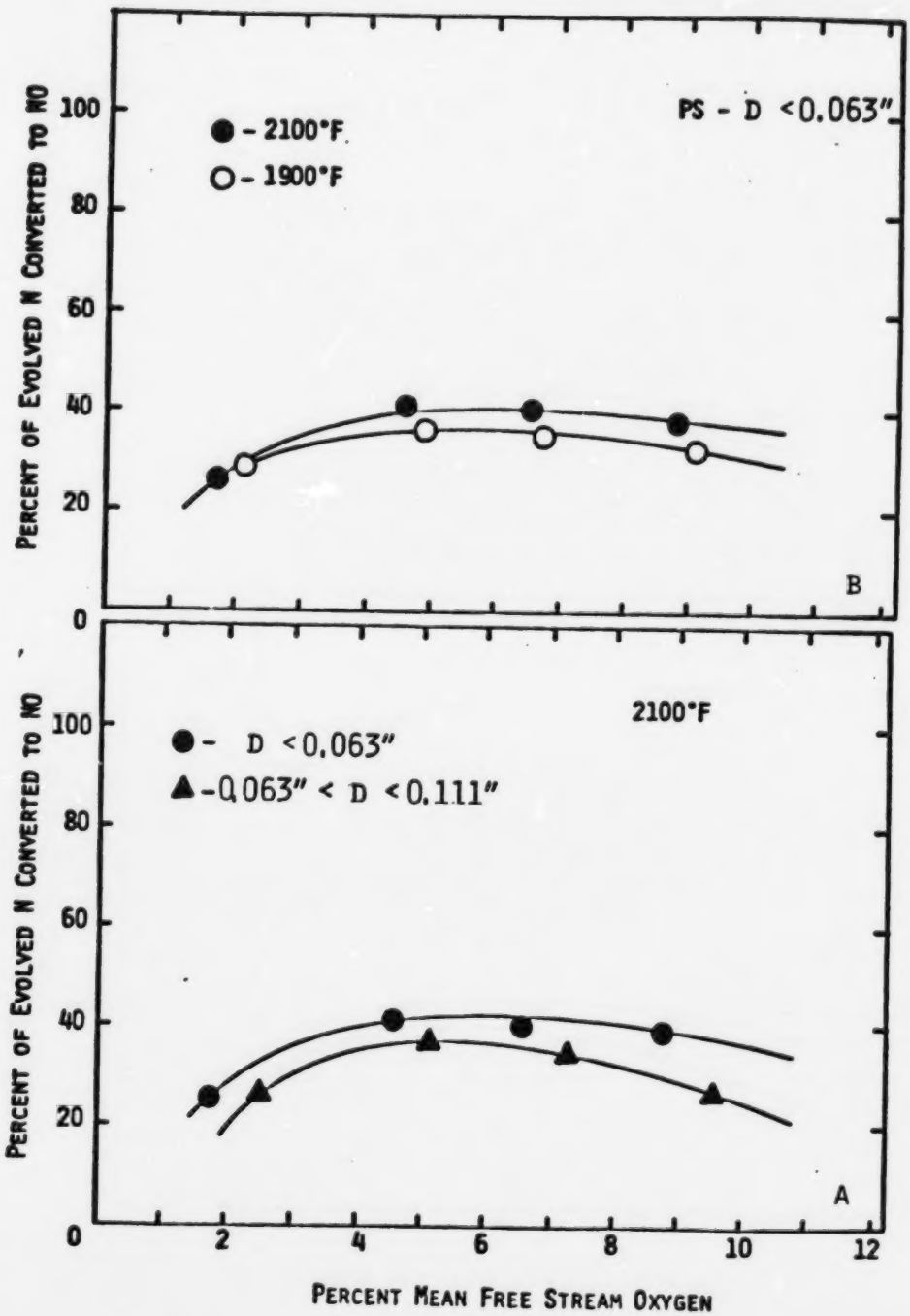


Figure 5. Influence of Combustion Conditions (Virginia Coal, Tower Furnace)

mean-free-stream oxygen, except at very low O_2 levels. The smaller particles exhibited slightly higher fuel nitrogen conversions (Figure 5A) and this agrees with pulverized coal results on much smaller particles.[11] Decreasing particle size is known to increase the volatile nitrogen evolution rate (Figure 4) and believed to enhance the probability of O_2 availability for XN (e.g., NH_3, HCN) oxidation. (The importance of intraparticle XN reduction reactions is decreased as particle size decreases.)[12] Bulk gas temperature had relatively little effect on fuel nitrogen conversion (Figure 5B).

The suspension phase results demonstrated that only particles of less than 0.1 in. diameter undergo significant reaction in the overthrow phase and that, overall, fuel nitrogen is evolved at approximately the same rate as carbon is oxidized. The conversion of the evolved nitrogen is relatively high (ca. 40%) and depends primarily on local oxygen availability. Particle size and thermal environment effects are of second-order importance.

FIXED-BED COMBUSTION

The fixed-bed furnace was utilized to study the formation and reaction of nitrogenous intermediates in proximity to the fuel bed. A Utah bituminous coal and a North Dakota lignite (Table 1) were fired under similar thermal and stoichiometric conditions.

Figure 6 illustrates the apparent fuel nitrogen conversion to NO for both fuels (NO formation by thermal fixation of N_2 was neglected). Overall stoichiometry was held constant (Utah at SR_T = 1.39, North Dakota lignite at SR_T = 1.45), and bed-region stoichiometry was varied. Under locally excess-air conditions the bituminous coal and lignite exhibited maximum fuel nitrogen conversions of approximately 16.5 and 13%, respectively. By contrast, the small particle (0.063 in.) suspension burning (Utah coal) showed a fuel nitrogen conversion of approximately 40%.

Both fuels showed a significant decrease in nitrogen conversion with decreasing bed-region stoichiometry. A weak minimum in nitrogen conversion was achieved for the Utah coal at a bed stoichiometry of 0.65. Both fuels showed a minimum fuel nitrogen conversion of 6.3% under staged burning. The relative change in fuel nitrogen with decreasing bed-region stoichiometry was greater for the Utah coal than for the lignite. Similar behavior has been observed in pulverized coal studies;[13] the relatively low staged NO emissions (Utah coal) resulted from high volatile nitrogen yields which, under fuel-rich conditions, can be reduced to N_2 prior to second-stage air addition.

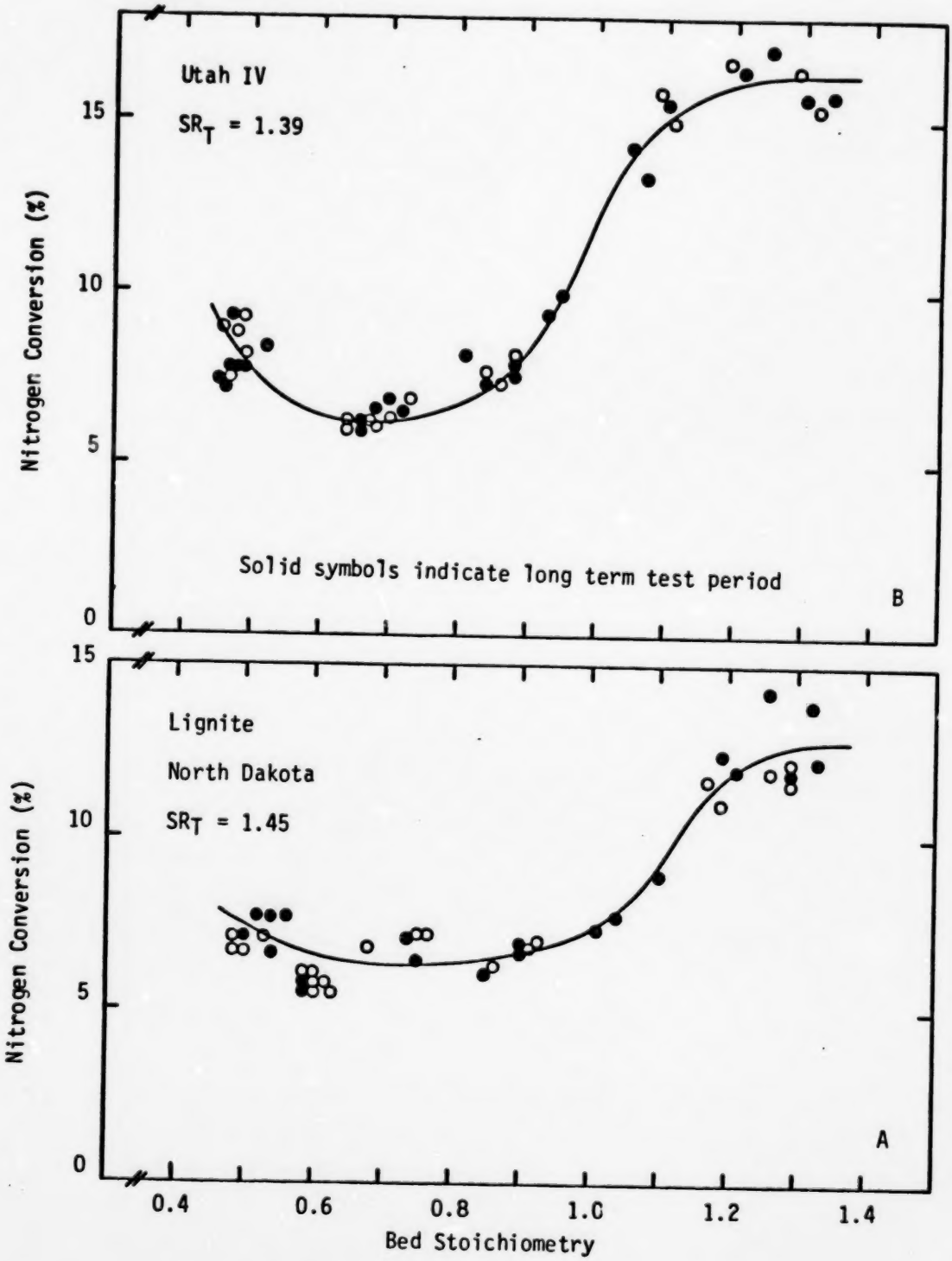


Figure 6. Relative Fuel Nitrogen Conversion to NO
(Fixed-bed Furnace)

Under extremely fuel-rich conditions (SR_{bed} < 0.60) the Utah coal (and to a lesser extent the lignite) exhibited increased NO emissions, a trend often observed under extreme staged burning in pulverized coal combustion.[8]

Figure 7 illustrates first-stage nitrogenous species for both fuels. These measurements were made at approximately 4 in. above the fuel bed. The secondary combustion air was added at 21 in.

For both fuels the first-stage NO decreased with decreasing bed-region stoichiometry. HCN and NH_3 concentrations increased dramatically when the local stoichiometry was reduced below 1.0 for the lignite and 0.90 for the Utah coal. Detailed gas chromatographic analyses were performed on the fuel-rich samples; however, no other significant nitrogenous intermediates were found. At the corresponding minimum in exhaust NO emissions, the first-stage nitrogenous species were dominated by HCN for the Utah coal (exhaust minimum at SR_{bed} = 0.65) and NH_3 for the lignite (exhaust minimum at SR_{bed} = 0.75). The predominance of HCN for bituminous coals and NH_3 for lignites is in agreement with first-stage pulverized coal measurements made by Chen et al.[13] , and is believed to result from differences in hydrocarbon volatiles evolved from the coal particles.

Under extremely fuel rich conditions (SR_{bed} < 0.65) the lignite showed a dramatic decrease in NH_3 formation while HCN remained constant and became the dominant fuel nitrogen species. The decreased gas-phase NH_3 contributed to lower TFN (TFN = NO + NH_3 + HCN) concentrations which resulted in low exhaust NO emissions. Thus, the lignite did not show the significant increase in NO emissions under heavily staged burning demonstrated by the Utah coal (Figure 6).

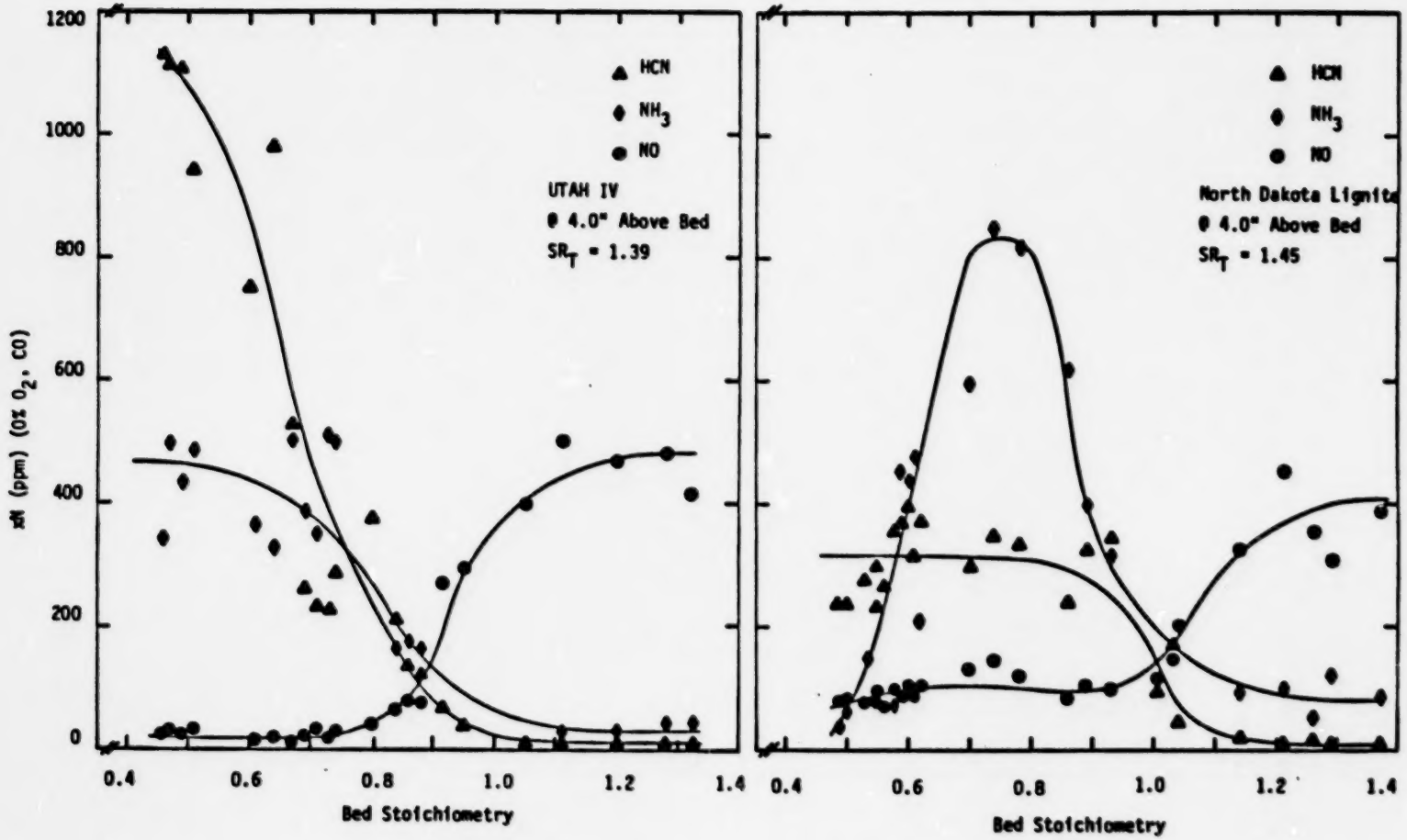


Figure 7. Intermediate Nitrogeneous Species
(Sampled at 4 in. above Bed, Fixed-bed Furnace)

The first stage NO remained fairly high (ca. 100 ppm) under sub-stoichiometric conditions for the lignite, while the Utah coal yielded NO concentrations of only 10 to 20 ppm. This may be a direct reflection of the high fuel oxygen content of the lignite.

Figure 8 illustrates the relationship between the total fixed nitrogenous species present in the first stage to NO in the exhaust. The data are presented in terms of TFN conversion as a function of TFN concentration in the first-stage (measured at 4 in.). No dependence of first-stage TFN conversion on original coal type was observed. The conversion was controlled by the local stoichiometry and the TFN concentration.

The excess-air-bed TFN concentrations (solid symbols) were dominated by first-stage NO and remained fairly constant at approximately 500 ppm; however, the TFN conversion for these data decreased with decreasing bed-region stoichiometry. TFN concentrations measured under fuel-rich bed conditions (open symbols) were dominated by HCN and NH_3, and these conversions were inversely proportional to local concentration. There is no obvious dependence of TFN conversions on the locally predominant nitrogenous species (HCN or NH_3).

Overall, the fixed-bed results indicated that the combustion could be successfully staged and substantial NO reductions achieved. Minimum fuel nitrogen conversions were very low (5-15%), compared to the suspension phase conversions (25-45%). In-flame measurements indicated that a minimum in exhaust NO was observed because the decrease in first-stage NO formation with decreasing bed-region stoichiometry was ultimately compensated for by an increase in other oxidizable, gas-phase nitrogenous species, principally NH_3 and HCN. NH_3 was found to be the dominant fixed-nitrogen species at the

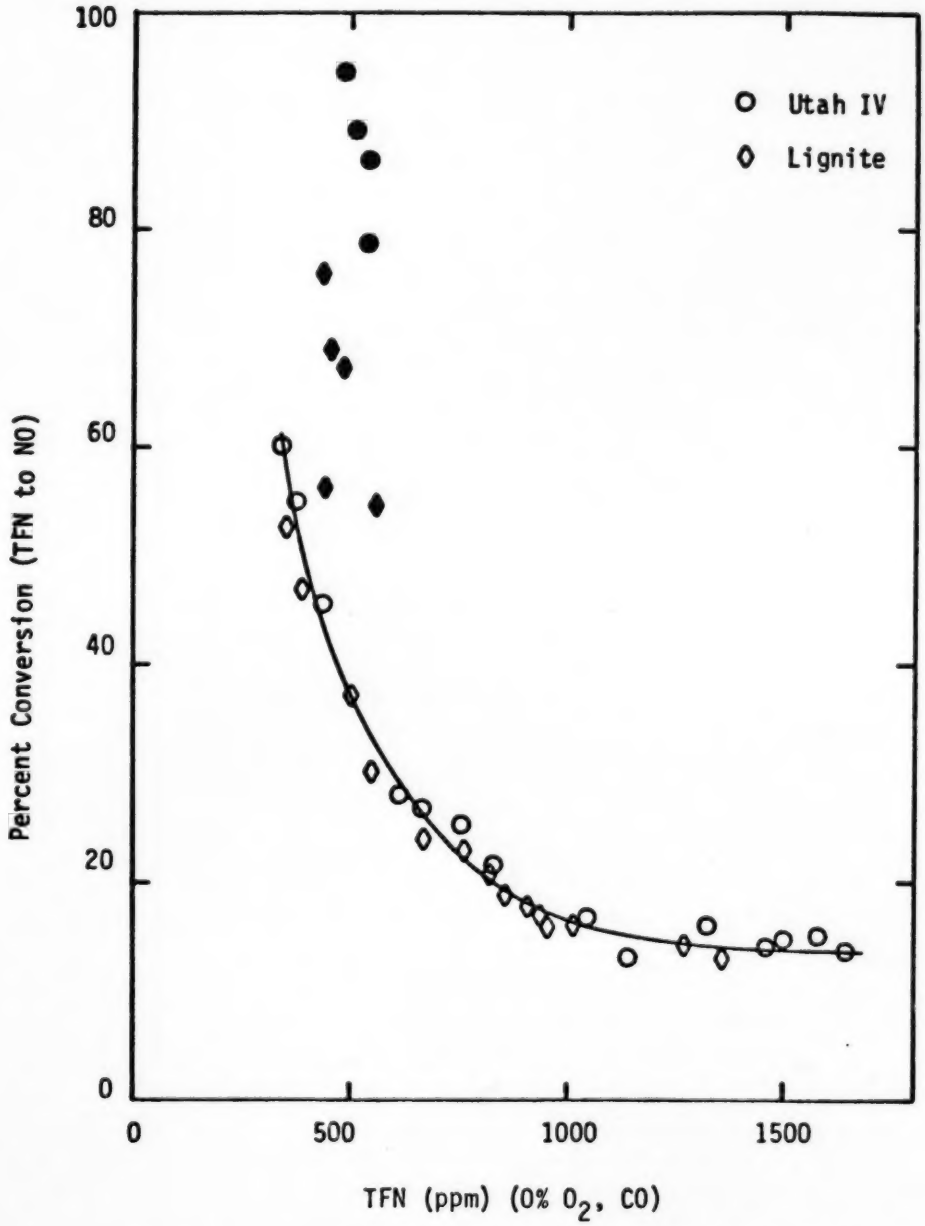


Figure 8. TFN Conversion to NO in Exhaust (TFN Measured at 4 in. above Bed, Overfire Air at 21 in., Fixed-bed Furnace)(Solid Symbols SR_{bed} >1.0, Open Symbols SR_{bed} <1.0).

optimum first-stage stoichiometry with the North Dakota lignite, while HCN was the dominant species with the Utah bituminous coal. Second stage conversion of the TFN was found to be inversely proportional to the TFN concentration but insensitive to differences in NH_3/HCN ratio.

MODEL SPREADER-STOKER STUDIES

Experiments were conducted in the model spreader-stoker furnace to investigate the coupling between the bed and suspension zone combustion and to assess the overall influence of the stoichiometry distribution and overfire-air height on NO formation and control. Initial validation tests were conducted to compare the pilot scale furnace with emissions data from full scale units. In full scale spreader-stoker systems, typical air distribution profiles employ 80 to 85% of the air as underfire air. The remaining air, overfire air, is split between two levels of jets, typically at approximate heights of 1.5 and 6 ft. above the grate. The effect of a change in the total air flow on a full scale furnace is illustrated in Figure 9 (solid lines) from the work of Langsjoen et al.[14]

NO emissions data from the model spreader-stoker unit, operated at typical field conditions, are also shown in Figure 9 (solid points). These tests were conducted with Utah coal at a firing rate of 470,000 Btu/hr. Overfire air represented 18% of the total air supplied to the furnace. The overfire air was introduced at 20 and 48 in. above the grate; the ratio of the air introduced at 48 in. to that at 20 in. was maintained constant at 1.15. The quantitative agreement between the pilot-scale model spreader-stoker emissions and the field testing data suggests that the pilot furnace appropriately simulates the combustion conditions which are of primary importance to NO formation.

In more extensive tests, the furnace air distribution (stoichiometry and overfire-air height) was varied. Total furnace stoichiometry was held constant at 1.45; the coal feed rate was 40 lb/hr (470,000 Btu/hr/ft^2).

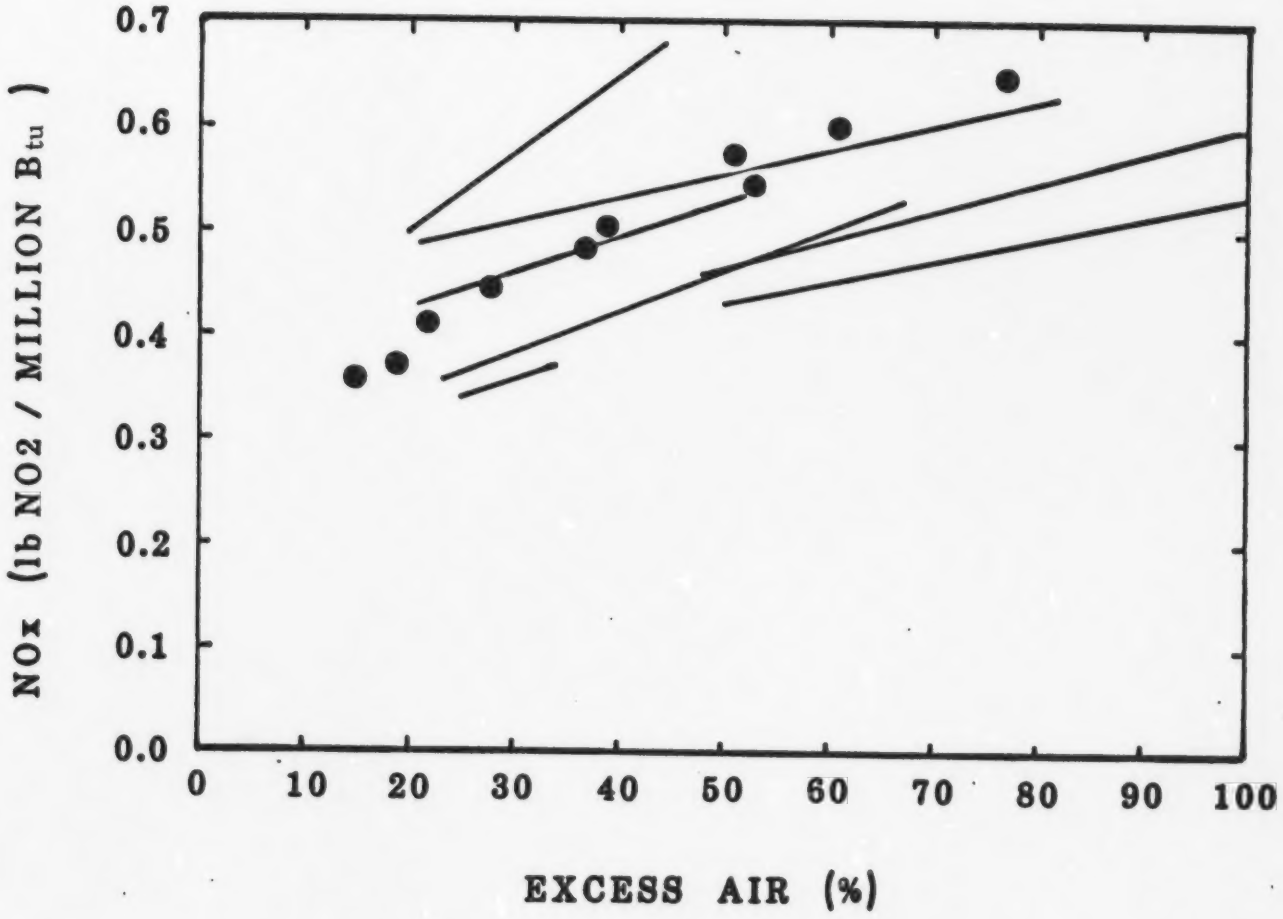


Figure 9. Comparison with Field Data (Solid Symbols MSF Data With Utah Coal, lines Ref. 14).

As in the fixed-bed studies, bed-region stoichiometry was found to be the principal parameter influencing NO emissions (Figure 10).

As the bed-region air/fuel ratio was reduced, the NO emissions decreased significantly. As the bed-zone stoichiometry was further reduced with the overfire air at 20 in. (below the spreader at 32 in), the emissions ultimately began to increase again just as in the fixed-bed experiments (Figure 6). However, the optimum stoichiometry for the model spreader-stoker furnace (SR_{bed} = 0.8) was much leaner than for the fixed-bed furnace. This suggests that the presence of the overthrow phase shifted the TFN distribution, probably by enhancing the homogeneous and heterogeneous destruction of NO in the first fuel-rich stage.

Figure 10B indicates the strong influence of overfire-air height on NO emissions during staged combustion. The height of overfire-air injection was varied from 7 to 72 in. above the grate; the level of coal injection (spreader) was at 32 in. Injecting the overfire air above the spreader enhances the probability that fuel N evolved from the small particles burning in the suspension phase will react to form N_2. In addition, increasing the overfire-air height increases the rich-zone residence time and allows TFN species leaving the fuel bed to decay further toward their low equilibrium levels.

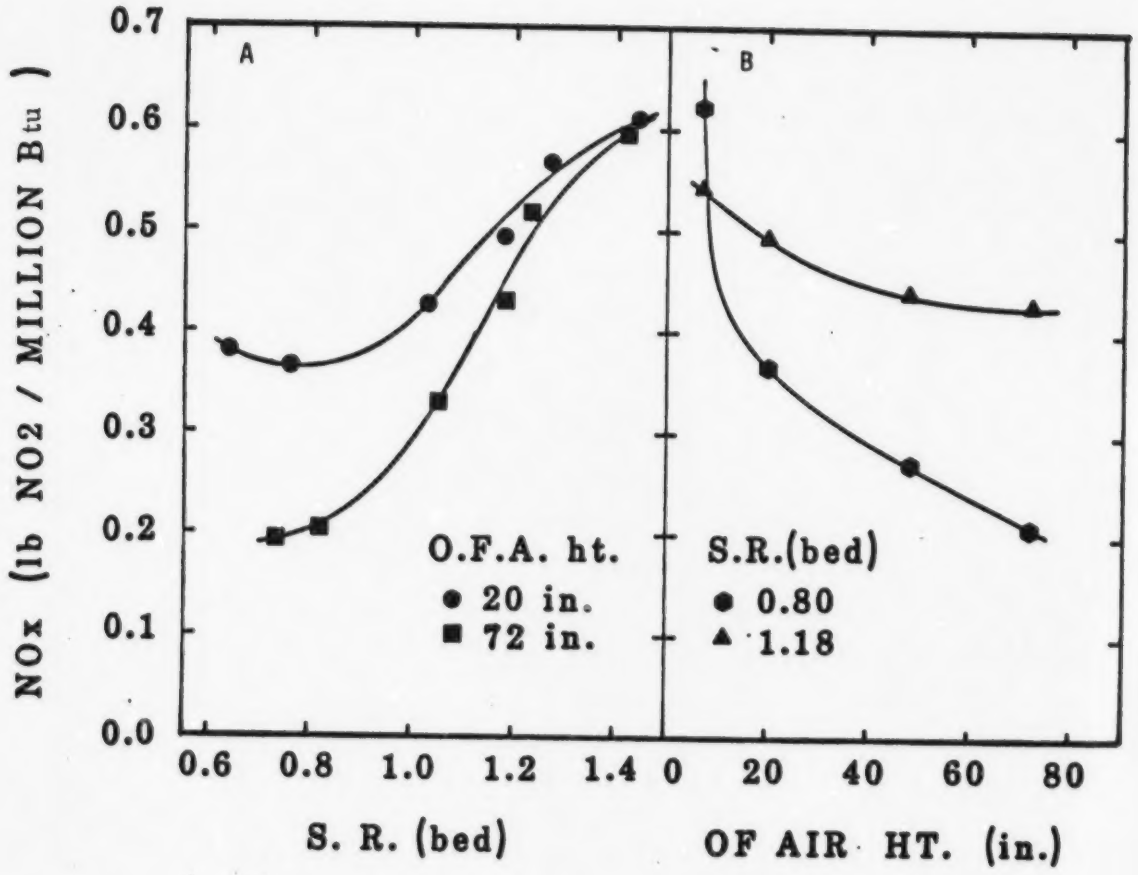


Figure 10. Influence of Air Distribution

CONCLUSIONS

This study indicates that NO emissions from spreader-stoker coal-fired furnaces are the result of relatively high conversions (25-45%) of the fuel nitrogen evolved from particles less than 0.11 in. in the suspension phase and low conversions (5 to 15%) of fuel nitrogen from fixed-bed combustion. In the suspension phase, nitrogen is evolved at approximately the same rate as carbon is oxidized, and the percentage conversion is relatively insensitive to reaction zone temperature and particle size. However, particle size dramatically influences the amount of fuel nitrogen evolved.

Local oxygen availability is the primary control parameter for both phases of the combustion. Substantial NO emission reductions were achieved when the fuel bed was operated under sub-stoichiometric conditions (fuel-rich). NO emissions decreased further when the suspension zone stoichiometry was reduced below 1.0 by moving the overfire-air injection location above the spreader. Minimum fuel nitrogen conversions of 6% were achieved, suggesting that staged combustion produces a positive synergism between the suspension phase and the bed combustion. The suspension zone provides sites for heterogeneous reduction of NO formed in the bed and the radical concentrations necessary to accelerate the decay of TFN formed in the bed. In addition, the low oxygen partial pressure decreases the conversion of nitrogen evolved in the suspension zone.

Thus, two types of control technology appear to be viable. First, the coal feed could be screened to remove the fines (particles of less than 0.1 in. diameter). This would essentially eliminate the high conversion of nitrogen evolved in the suspension zone and would result in an overall

emissions reduction of approximately 5 to 25% depending on the amount of fines normally present in the raw coal. In addition, it might result in a more permeable coal layer on the grate which would improve underbed air distribution. Alternatively, the primary overfire air injection port could be moved above the spreader and the suspension zone and bed-region could both be operated sub-stoichiometric. This could result in an emission reduction as large as 50%; however, before this concept can be applied commercially, the influence on other important combustion parameters (e.g., bed thickness, grate temperature, and ash characteristics) must be evaluated.

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